

Inferring black carbon content and specific absorption from Aerosol Robotic Network (AERONET) aerosol retrievals

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[1] Black carbon is ubiquitous in the atmosphere and is the main anthropogenic absorbing particulate. Absorption by black carbon is thought to be comparable to the cooling associated with sulfate aerosols, although present-day satellites are incapable of obtaining this measurement, and model estimates are highly uncertain. More measurements of black carbon concentration are necessary for improving and validating transport and general circulation models. The Aerosol Robotics Network (AERONET) of 180 worldwide radiometers offers an opportunity to obtain these measurements. We use the Maxwell Garnett effective medium approximation to infer the column-averaged black carbon concentration and specific absorption of AERONET retrievals at 46 locations. The yearly averaged black carbon column concentrations exhibit the expected regional dependence, with remote island locations having values about an order of magnitude lower than the continental biomass burning locations. The yearly averaged black carbon specific absorption cross section is consistent with other measured values, $9.9 \text{ m}^2 \text{ g}^{-1}$ for 19,591 retrievals, but varies from 7.7 to $12.5 \text{ m}^2 \text{ g}^{-1}$. We attribute this variability to the details of the size distributions and the fraction of black carbon contained in the aerosol mixture. We also used the Maxwell Garnett equations to parameterize the imaginary refractive index with respect to the black carbon volume fraction, enabling simple but accurate absorption estimates for aerosol mixtures when the black carbon fraction and size distribution is known. The black carbon concentrations that we derive from AERONET measurements correctly describe the radiance field and represent an alternative to absorption optical thickness in the link between models and AERONET measurements.

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1. Introduction

[2] It is often reported that aerosols have slowed global warming by scattering incident solar radiation back into space, and that the impact of anthropogenic aerosols is a global cooling comparable in magnitude (but opposite in sign) to the warming associated with anthropogenic infrared-active gases [Charlson *et al.*, 1991, 1992; Penner *et al.*, 1992; Harshvardhan, 1993; Schwartz, 1996; Delene and Ogren, 2002]. While scattering is the predominant radiative effect of aerosols at shortwave wavelengths (i.e., less than $4 \mu\text{m}$), significant absorption by aerosols occurs at these wavelengths as well [Lioussse *et al.*, 1996; Haywood and Shine, 1997; Schult *et al.*, 1997; Myhre *et al.*, 1998; Penner *et al.*, 1998; Jacobson, 2001]. The most recent report from the Intergovernmental Panel on Climate Change (IPCC) [IPCC, 2001] estimates that absorption by black carbon aerosols can reduce the direct radiative impact of

sulfate aerosols by 50–100%. Aerosol absorption is significant enough that the radiative impact of aerosols at the top of the atmosphere could change in sign from cooling to warming in regions of highly absorbing aerosols [Chylek and Coakley, 1974; Charlock and Sellers, 1980; Haywood and Shine, 1995]. This is especially important when aerosols are located over highly reflective surfaces such as snow or clouds [Haywood *et al.*, 1997]. Absorbing aerosols may also be responsible for a second indirect effect, whereby clouds evaporate more rapidly because of absorbing haze and cloud condensation nuclei [Ackerman *et al.*, 2000]. A recent study also indicates that such aerosols reduce the albedo of ice and snow and increase melt rates [Hansen and Nazarenko, 2004].

[3] Absorption by nondust aerosols in the atmosphere at wavelengths less than $4 \mu\text{m}$ is mainly caused by the graphitic form of carbon [Rosen *et al.*, 1978, 1982]. This highly absorbing particulate is often called carbon black, soot, elemental carbon, or black carbon in the atmospheric literature. Differences between these species do exist, however. Carbon blacks refers to commercially available spherical particles created in a controlled environment. These

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particles are composed of concentric graphite platelets with diminishing graphitic order near the center [Hess and Herd, 1993]. Their commercial availability simplifies the characterization of their physical and optical properties and these properties are often extrapolated to characterize particulate carbon found in atmospheric soot. Atmospheric soot is produced by the same mechanism as carbon blacks (incomplete combustion), but it also contains a variety of impurities [Novakov, 1982; Bansal and Donnet, 1993]. For instance, diesel exhaust soot has a much lower carbon content than commercial carbon blacks ($\sim 45\%$ versus $\sim 95\%$) even though they both exhibit very similar primary particle sizes and graphitic internal structure [Clague et al., 1999]. The term elemental carbon implies a purity of substance and is generally reserved for thermal analysis of light absorbing carbon, the philosophy being that the various impurities volatilize at a lower temperature than the graphitic component. The term black carbon is often reserved for optical measurements of light absorbing carbon (A. D. A. Hansen, 2003; see www.mageesci.com), which can show excellent agreement with thermal elemental carbon measurements [Moosmüller et al., 1998]. We adopt the term black carbon to refer to nonorganic particulate carbon in the atmosphere with the understanding that the optical and physical properties that we obtain from the literature may be associated with carbon blacks or soot.

[4] Current satellite technology does not include aerosol absorption measurements, so global estimates must be obtained from transport models and general circulation models (GCMs). The models are typically initialized with gridded emission inventories of all known significant aerosol sources and assumed size distributions. The aerosols are then transported across the globe, changing in mass and optical properties as the modeled relative humidity changes. Removal occurs by wet and dry deposition. The carbon emissions inventories used to initialize the models are highly parameterized and created on the basis of multiple sparse data sets (such as fuel use inventories and emission factors). The resulting inventories are uncertain by at least a factor of 2 and this uncertainty is carried forward to the model output [Bond et al., 1998; Cooke et al., 1999; Streets et al., 2001].

[5] Complicating matters further, aerosol models often assume external mixtures so that the hygroscopic growth and radiative impact of each component may be considered separately [Kinne et al., 2003], although internal mixtures of black carbon has been explored in at least one model [Jacobson, 1997]. (The external mixture assumption presumes that each aerosol particle contains a single species, whereas multiple species are contained within each particle in an internal aerosol mixture.) Measurements show that black carbon and sulfate concentrations are highly correlated, indicating that these two species may be internally mixed [Pinnick et al., 1993; Krivacsy et al., 2001]. This hypothesis is confirmed with high-resolution scanning electron microscopy images [Ebert et al., 2002] and is consistent with the deposition rates required by models to remove carbon from the atmosphere; dry deposition is too slow, so modelers regularly assume a hydrophobic to hydrophilic conversion for wet removal of carbon [Collins et al., 2001]. Such a conversion would be unlikely in reality unless the carbon was already mixed with a hydrophilic substance. Sulfate is extremely hydrophilic and created in the same combustion processes as soot, providing a plausible physical substrate for this parameterization.

7. Conclusions

[80] Continuous worldwide measurements of black carbon concentrations are required to improve the current gridded carbon emissions inventories and transport models. Black carbon specific absorption measurements are also desirable for relating the modeled microphysics to aerosol optical properties. We have developed a technique for retrieving both the black carbon column concentration and specific absorption from the worldwide AERONET database.

[81] Particulate carbon absorbs visible radiation more efficiently when it is contained within a host aerosol (i.e., internal versus external mixing) but the increase in efficiency is not uniform for all internal mixtures of aerosols. We calculated the range of possible specific absorptions for internal mixtures of black carbon using nine climatological size distributions, and found a factor of 2 or more variability for black carbon fractions typical of atmospheric aerosols. The results are highly dependent upon the volume fraction of black carbon but independent of specific combustion processes, as we used a single refractive index for black carbon in this study. This indicates that a single number can not be used to accurately convert thermal black carbon concentration measurements to absorption (and vice versa) without knowledge of the aerosol size distribution and fraction of black carbon.

[82] We used the Maxwell Garnett effective medium approximation to infer the column-averaged concentration and specific absorption of black carbon associated with the AERONET retrievals at 46 locations. The yearly averaged black carbon column concentrations that we found are comparable to typical measured concentrations if a 1 km boundary layer is assumed: $0.22\text{--}0.28\ \mu\text{g m}^{-3}$ at remote island locations, $0.96\text{--}3.47\ \mu\text{g m}^{-3}$ in continental regions, and $2.7\text{--}3.7\ \mu\text{g m}^{-3}$ in biomass burning locations (see Table 2). Likewise, the specific absorptions we infer at these locations are consistent with other reported values. The ocean sites have a higher specific absorption (averaging $11.3\ \text{m}^2\ \text{g}^{-1}$ for 2200 retrievals) and the biomass burning sites have a lower specific absorption ($8.9\ \text{m}^2\ \text{g}^{-1}$ for 3942 retrievals) than the continental sites ($9.9\ \text{m}^2\ \text{g}^{-1}$ for 13,449 retrievals) because of the inverse relationship of specific absorption to black carbon concentration.

[83] We also used the Maxwell Garnett effective medium approximation to parameterize the imaginary refractive index with respect to the black carbon volume fraction, enabling simple but accurate absorption estimates for aerosol mixtures when the black carbon fraction is known. The parameterization indicates that the mixture imaginary refractive index is sensitive to the real refractive index of the host aerosol, but not to additional inclusions. We compared our parameterization to the volume-averaged mixing approximation and found a 13–30% high bias for volume-averaged imaginary refractive index and the host aerosols tested; this corresponds to a 13–23% low bias in the inferred black carbon concentration if volume averaging is substituted for the Maxwell Garnett equations in our retrieval. The high bias for the imaginary refractive index produced by the volume mixing approximation is very similar to an “inverted Maxwell Garnett” mixture consisting of a black carbon host aerosol with nonabsorbing inclusions.